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Citation: The Journal of Chemical Physics **135**, 047101 (2011); doi: 10.1063/1.3615517
View online: http://dx.doi.org/10.1063/1.3615517
View Table of Contents: http://scitation.aip.org/content/aip/journal/jcp/135/4?ver=pdfcov
Published by the AIP Publishing

Comment on "On the role of dissipation on the Casimir-Polder potential between molecules in dielectric media" [J. Chem. Phys. 133, 164501 (2010)]

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(Received 24 February 2011; accepted 10 May 2011; published online 28 July 2011)

[doi:10.1063/1.3615517]

The problem of the Casimir force for bodies immersed in a third medium was first solved by Dzyaloshinskii and Pitaevskii,[1] and various aspects of related problems have attracted much attention in recent years. For instance, an expression has been obtained for the van der Waals (or Casimir-Polder) interaction between two molecules in a dispersive and dissipative dielectric medium:[2–4]

$$W(|\mathbf{r}_a-\mathbf{r}_b|) = -\frac{\hbar}{2\pi}\int_0^\infty du\alpha^a(iu)\alpha^b(iu)G_{ij}(\mathbf{r}_a,\mathbf{r}_b,iu)G_{ji}(\mathbf{r}_a,\mathbf{r}_b,iu), \tag{1}$$

where $\alpha^a(iu)$ and $\alpha^b(iu)$ are the electric polarizabilities, evaluated at imaginary frequency, of atoms a and b, $\mathbf{r}_a$ and $\mathbf{r}_b$ denote the positions of the atoms, and summation over repeated indices is implied. The Green dyadic $\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)$ satisfies

$$\left[\nabla\times\nabla\times-\frac{\omega^2}{c^2}\epsilon(\mathrm{r},\omega)\right]\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)=\mathbf{1}\delta(\mathbf{r}-\mathbf{r}',\omega), \tag{2}$$

together with the usual boundary condition at infinity, and is given in the case of a homogeneous dielectric medium with complex permittivity $\epsilon(\omega)$ and refractive index $n(\omega)$ by[5]

$$G_{ij}(\mathbf{r},\mathbf{r}',\omega)=\left(1+\frac{1}{k_m^2}\nabla\nabla\right)\frac{e^{ik_m|\mathbf{r}-\mathbf{r}'|}}{|\mathbf{r}-\mathbf{r}'|}, \tag{3}$$

where $k_m=\sqrt{\epsilon(\omega)}\omega/c=n(\omega)\omega/c$. One obtains

$$G_{ij}(\mathbf{r},\mathbf{r}',\omega)=\left[\delta_{ij}-R_iR_j/R^2-(\delta_{ij}-3R_iR_j/R^2)\times\left(\frac{1}{k_m^2R^2}-\frac{i}{k_mR}\right)\right]\frac{e^{ik_mR}}{R}\quad(R=|\mathbf{r}-\mathbf{r}'|), \tag{4}$$

which results in the familiar formula for the electric field at all distances from an electric dipole. The van der Waals interaction derived by Rodriguez and Salam (RS) (Ref. 6) has the same form as (1), but their Green function differs from (4) and, therefore, the interaction they obtain differs from that of Ref. 2. We argue here that the formulas they use for the Green function and the electric field violate basic requirements of causality and unitarity, and therefore that RS do not obtain a valid van der Waals interaction.

Consider first the Green dyadic. The well-known expression (4) is analytic in the upper half I_+ of the complex frequency plane, as required by causality,[7] since **G** simply relates the electric field to its source. The Green function in Eq. (15) of RS, however, violates this basic requirement. Specifically, the first term in the second equality of their expression (15) is proportional to the imaginary part of the refractive index $n(\omega)$, which, unlike $n(\omega)$, is not analytic in I_+.[8] Unlike (4), furthermore, the Green function of RS is not retarded; it has a term without a factor $\exp(ik_mR)$, corresponding to an instantaneous, static electric field. RS obtain as in Ref. 2 the correct $|\mathbf{r}_a-\mathbf{r}_b|^{-7}$ dependence of the interaction energy at large separations, the so-called "retarded" van der Waals interaction, but this does not contradict the fact that their Green function is not retarded in the usual sense, as the static part of their Green function varies as R^{-3} and does not contribute to the large-separation van der Waals interaction.

The issue of retardation bears on the RS conclusion that the van der Waals interaction persists even in the limit of large absorption: because it has no factor $\exp(ik_mR)$, the nonretarded part of their **G** does not decay with R as $\exp(-\mathrm{Im}[k_m]R)$. RS associate this part of **G** with the longitudinal component of the electric field, which, like the transverse component, is not retarded.[9] They appear to ignore the obvious fact that it is the full, *retarded* electric field, longitudinal plus transverse, that is physically meaningful[9,10] and mediates the interaction, and that this field from any source in the medium is retarded.

RS claim that their Green function was "derived by explicitly accounting for the effect of absorption in the medium," but what they in fact do is *infer* their **G** by calculating the interaction using perturbation theory with the electric dipole interaction [see their Eq. (6)]

$$\hat{H}_{\mathrm{int}}=-\hat{\mathbf{p}}_a\cdot\hat{\mathbf{E}}(\mathbf{r}_a)-\hat{\mathbf{p}}_b\cdot\hat{\mathbf{E}}(\mathbf{r}_b), \tag{5}$$

where $\hat{\mathbf{p}}$ is the electric dipole moment operator and the electric field operator $\hat{\mathbf{E}}$ is given in their Eq. (1). (We use circumflexes to denote operators.) In other words, their **G**, and all the conclusions that RS draw from it, is based on their expression for $\hat{\mathbf{E}}$, which we now argue is invalid.

For frequencies ω_k at which absorption can be ignored, the electric field operator has the form[11]

$$\hat{\mathbf{E}}(\mathbf{r},t)=i\sum_{\mathbf{k}\lambda}\left(\frac{2\pi\hbar\omega_k}{V}\right)^{1/2}[(n_k)^{-1/2}\hat{a}_{\mathbf{k}\lambda}(t)\mathbf{e}_{\mathbf{k}\lambda}e^{i\mathbf{k}\cdot\mathbf{r}}-\mathrm{h.c.}] \tag{6}$$

in the familiar notation in which $\hat{a}_{\mathbf{k}\lambda}$ and $\hat{a}^{\dagger}_{\mathbf{k}\lambda}$ are annihilation and creation operators, respectively, for the field mode with propagation vector $\mathbf{k}$ and polarization λ; $\mathbf{e}_{\mathbf{k}\lambda}$ is a polarization unit vector, V is a quantization volume, and n_k is the (real) refractive index at frequency ω_k. For simplicity, we have written the field for the case in which dispersion is negligible. Now in writing their $\hat{\mathbf{E}}$ RS assert that, in the case of an absorbing medium, all one needs to do is replace $\hat{a}^{\dagger}_{\mathbf{k}\lambda} n_k^{-1/2}$ in (6) by $\hat{a}^{\dagger}_{\mathbf{k}\lambda} n^{*-1/2}_k$, thus maintaining Hermiticity, and allow $\mathbf{k}$ to be complex. They cite for support of this the work in Ref. 12, but in Ref. 12 it was assumed that the refractive index at frequencies of interest is purely real, i.e., that *the medium is non-absorbing*. The generally accepted expression for the quantized electric field in a dispersive and absorbing homogeneous dielectric medium[13] is very different from that employed by RS. In particular, the $\hat{\mathbf{E}}$ assumed by RS decays exponentially with propagation distance from a point $\mathbf{r} = 0$ that they regard as "the point of creation of the field." It is not clear what defines this point of creation; in any event their expression for the field implies Beer-law decay of the electric field *operator*. This implies that canonical commutation relations for electromagnetic field operators decay to zero from this point of creation, thus violating another fundamental requirement (unitarity) that such commutation rules be preserved.

RS misinterpret a conclusion of Ref. 2 when they state that absorption in the medium "must yield a result for the dispersion potential that is different than if it were ignored altogether, unlike as claimed in Ref. 2." We made no such claim. We only showed that one could derive the interaction without *explicit* account for absorption, as long as the correct analytical properties of the permittivity are recognized in order to be able to express as in Eq. (1) the interaction in terms of imaginary frequencies, *for which the permittivity is real regardless of absorption*. This circumstance is well known in the context of the Lifshitz theory, as was discussed in our paper. In particular, the remarks after Eq. (46) of Ref. 2 make it abundantly clear, in our opinion, that the various van der Waals interactions considered there are in fact affected by absorption.

While we are unaware of any experimental evidence that supports or refutes the results of Ref. 2 or of RS, the inconsistencies of the latter with some basic principles cast very serious doubt on their validity.

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[1]I. E. Dzyaloshinskii and L. P. Pitaevskii, Sov. Phys. JETP **9**, 1282 (1959). See also L. P. Pitaevskii, "On the problem of van der Waals forces in dielectric media," in *Casimir Physics*, Lecture Notes in Physics, edited by D. A. R. Dalvit, P. W. Milonni, D. C. Roberts, and F. S. S. Rosa (Springer-Verlag, Berlin, 2011); e-print arXiv:1011.5591v1 [cond-mat.stat-mech].

[2]S. Spagnolo, D. A. R. Dalvit, and P. W. Milonni, Phys. Rev. A **75**, 052117 (2007).

[3]Results of the same form were obtained by M. S. Tomaš, J. Phys. A **39**, 6785 (2006).

[4]We consider here only electrically polarizable guest atoms and host media.

[5]H. Levine and J. Schwinger, Commun. Pure Appl. Math. **3**, 355 (1950).

[6]J. J. Rodriguez and A. Salam, J. Chem. Phys. **133**, 164501 (2010).

[7]Analyticity of the Green function in I_+ follows from the same (Kramers-Kronig) causality arguments that demand that the dielectric permittivity and the refractive index have this property. See, for instance, J. D. Jackson, *Classical Electrodynamics*, 2nd ed. (Wiley, New York, 1975), p. 309. In the case of the dyadic Green function for a dielectric medium, analyticity in I_+ has been emphasized by other authors. See, for instance, A. Narayanaswamy and G. Chen, J. Quant. Spectrosc. Radiat. Transf. **111**, 1877 (2010).

[8]A simple example: $n(\omega) = 1 + A/[\omega_0^2 - \omega^2 - i\beta\omega]$, $\beta > 0$, is analytic in I_+, but $\mathrm{Im}[n(\omega)] = A\beta\omega/[(\omega_0^2 - \omega^2)^2 + \beta^2\omega^2]$ is not.

[9]See, for instance, P. W. Milonni, *The Quantum Vacuum. An Introduction to Quantum Electrodynamics* (Academic, San Diego, 1994), Sec. 4.6, and references therein. The nonretarded part of the transverse Green function is cancelled by the longitudinal Green function, so that the complete Green function is properly retarded. These properties of the Green function used in Ref. 2 in the case of an absorbing medium can be seen explicitly in Appendix A of S. M. Barnett, B. Huttner, R. Loudon, and R. Matloob, J. Phys. B **29**, 3763 (1996).

[10]For an interesting discussion of this point, and its implications for retardation, see J. D. Jackson, Eur. J. Phys. **31**, L79 (2010), where it is remarked that "The longitudinal and transverse components [of the electric field] have no separate physical realities; only the sum is physical."

[11]See, for instance, P. W. Milonni, J. Mod. Opt. **42**, 1991 (1995) and references therein.

[12]P. D. Drummond and M. Hillery, Phys. Rev. A **59**, 691 (1999).

[13]B. Huttner and S. M. Barnett, Phys. Rev. A **46**, 4306 (1992). In their paper RS do not cite this or any other work on field quantization in dissipative media.